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The absence of crystallization on supercooling in a single component system of particles interacting through the harmonic-repulsive potential. A possible formation of a quasicrystal at a different value of the density. Investigations with direct molecular dynamics simulations.

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In this note, we report about two, as it seems to us, rather unusual observations made in molecular dynamics simulations of the single component systems of particles interacting through the harmonic-repulsive pair potential in 3D. In particular, at some densities, we observed deeply supercooled liquid states which did not exhibit crystallization in rather long MD runs. This observation is unusual because usually liquids formed by particles of only one type rather readily crystallize on supercooling. At a different value of the density we observed crystallization of the liquid into a state formed by chains (or lines) of particles which organize themselves into columns in such a way that each column is formed by seven lines of particles. Alternatively, one can think that each such column is formed by three alternating helical coils. In our view, it is possible that the overall structure that crystallizes from the liquid at this density is quasicrystalline. If it is indeed the case, then this observation is unusual because, as far as we understand, it is not expected, at present, that quasicrystalline structures can form in such a simple potential as harmonic-repulsive potential. Investigations related to the presumable phase diagram of the particles interacting through the harmonic-repulsive potential have been reported in [J. Chem. Phys. **134**, 044903 (2011)]. Our results appear to be at odds with some of the results presented there.

I. INTRODUCTION

If spherically symmetric pair potentials are used to model the behavior of the atomic systems then these potentials usually have steep repulsion at short distances and they diverge at zero separation between the model particles. The most common example is the Lennard-Jones potential often used to model the properties of the gases, liquids, and solids of the inert atoms. If crystal structures arise in the simulations with such hard-core pair potentials then these structures are usually Face Centered Cubic (FCC) or Body Centered Cubic (BCC) lattices. [1–6]

Over the last thirty years significant attention has been paid to modeling of the soft matter systems [7–13]. Some soft matter systems consist of star polymers or dendrimers, or micelles, or microgels in solutions. These mesoscopic particles may have approximately spherical shape and they can be modeled with spherically symmetric pair potentials [8–13].

The interactions between particles in the soft matter systems are quite different from the interactions between atoms. The corresponding modeling pair potentials may have only repulsive part (no attraction between the particles) and this repulsive part usually is much softer than the repulsive part met in the atomic interactions [7–13]. Moreover, complete overlap of the mesoscopic particles is sometimes possible and correspondingly, in modeling of such systems, there is only a finite energy penalty for the complete particles' overlaps, i.e., some modeling potentials have finite value at zero separation distance between the particles [7–13]. At present, it is well known that the

systems of particles interacting through soft potentials can form rather unusual structures in comparison to the structures formed by particles interacting through simple spherically symmetric atomic “hard-core” pair potentials [7–13].

Discovery of quasicrystals lead, in particular, to the systematic studies of the single component systems consisting of the particles interacting through the spherically-symmetric pair potentials whose shape is more complex than the shape of the simple “traditional” pair potentials [14–21]. The goal of the related studies is often to clarify the relationship between the shape of the potential and the structural/dynamic properties of the systems of particles interacting through such potentials.

Finally we mention two other research directions for which the results described in this note might be of interest. One direction concerns the investigations of the “unusual” interaction pair potentials that lead to the non-crystalline ground states [22–24]. Another research direction concerns the issue of the reverse engineering, i.e., the intent to design particles and their interactions in a way that would lead to the desirable structural and other properties [25–29].

In this note we report about some of our investigations of the systems of identical particles interacting through the harmonic-repulsive pair potential:

$$\phi(r) = \epsilon \left(1 - \frac{r}{\sigma}\right)^2 \theta\left(1 - \frac{r}{\sigma}\right), \quad (1)$$

where $\theta(x)$ is the Heaviside step function, while ϵ and σ determine the energy and length scales of the potential.

We note here that the phenomenon of clustering is not expected to occur for the systems of particles interacting

through potential (1)[9, 30].

Behavior of the particles interacting through potential (1) has been investigated before in several different contexts in a number of previous publications [10, 31–36]. In particular, in Ref.[34] the phase diagram of the system has been constructed. Our paper also concerns the issue of the system’s phase diagram. The point is that some of our results appear to be in disagreement with the results presented in Ref.[34]. See also Ref.[35].

The method used in Ref.[34] to construct the phase diagram consists of three steps. On the first step the set of the possible crystal structures is assumed. On the second step the set of the considered crystal structures is narrowed through the check of their stabilities at the density of interest (at low temperature) using the method of dissipative particle dynamics (after all, it is a particular method of molecular dynamics). On the third step the phase diagram of the system is constructed through calculations and comparisons of the free energies for the considered crystal structures. We note that the analysis implemented in Ref.[34] previously was also implemented in Ref.[37] (with the most traditional MD) in order to construct the phase diagram of the system of particles interacting through the repulsive Hertzian potential.

We, in our approach, acted in a less sophisticated way. Originally we became interested in this potential because we wanted to address a particular point described in Ref.[38, 39]. We used LAMMPS molecular dynamics program [40, 41] and traditional MD in order to produce liquid state through melting of the initial FCC crystal. The initial structure is of no importance after the melting. Then we cooled the liquid produced in this way. At certain particles’ densities we observed rather unusual behaviors of the systems. We performed MD simulations at a number of different densities. In this note we report about the two unusual results obtained at the densities $\rho_o\sigma^3 = 2.904$, $\rho_o\sigma^3 = 4.400$, and $\rho_o\sigma^3 = 4.500$.

In particular, at the density $\rho_o\sigma^3 = 2.904$, we observed crystallization of the liquid into a state that could be quasicrystalline. In this state the particles organize into linear chains which, in their turn, align into columns in such a way that each column is formed by seven linear chains of the particles. Alternatively, one can think about each seven-column as of being formed by three helical coils. It appears that the described structure is stable in some range of temperatures.

At the densities $\rho_o\sigma^3 = 4.400$, and $\rho_o\sigma^3 = 4.500$ we did not observe crystallization of the liquid into any crystal despite rather long molecular dynamics runs at a number of different supercooled liquid temperatures. This observation is unusual because usually single component systems of particles easily crystallize on supercooling.

The note is organized as follows. In section (II) the details of MD simulations and the methodology of the structure analysis are described in details. In section (III) the results of the MD simulations and the structure analysis are presented. We conclude in section (IV).

II. DETAILS OF MD SIMULATIONS AND DATA ANALYSIS

We performed molecular dynamics simulations (MD) using the LAMMPS program [40, 41]. The simulations were performed using the Lennard-Jones (LJ) units [42]. This choice of units determines the energy scale: if the value of the harmonic repulsive potential (1) at zero separation is equal to one, i.e., $\epsilon = 1$ then ϵ corresponds to the depth of the minimum of the LJ-potential in the LJ-units. If the length scale of the harmonic repulsive potential is equal to one, i.e., $\sigma = 1$ then this length corresponds to the particles’ diameter associated with the LJ-potential.

In the following all results will be presented in the LJ-units in accordance with the LAMMPS conventions, i.e., the temperature, T , and the Potential Energy per Particle (PEpP) will be measured in the units of ϵ . The time, t , will be given in the units of $\tau = \sqrt{\sigma^2 m / \epsilon}$ [42].

The magnitude of the time step was determined in the constant energy runs (NVE-ensemble) so that the total energy of the system is conserved with high precision (essentially no variation in the sixth digit of the value of the total energy per particle). The data were acquired in the constant temperature runs (NVT-ensemble) with Nosé-Hoover thermostat. The value of the time step varied in the interval between $\delta t \sim 0.001\tau$ for high temperatures $T \sim 0.015$ and $\delta t \sim 0.1\tau$ for nearly zero temperature $T \sim 0.000025$. The value of the dumping parameter associated with the Nosé-Hoover thermostat was chosen to be approximately equal to the 100 time steps.

Most of our simulations were performed on the systems containing 13500 particles in a cubic simulation box. Sometimes we used 16000 particles. The periodic boundary conditions were always implemented. In our simulations we did not notice size effects. Correspondingly, in the following discussions, we will not mention the sizes of the systems on which the data were obtained.

A. Data collection algorithm

At all densities we followed the same algorithm for the data collection.

1) At first, we generated a system at the required density as FCC lattice. It does not matter if this FCC lattice is stable or not at this density as on the next step the system was heated to a temperature at which it becomes a liquid. The fact that the system is in a liquid state was monitored using the Pair Density Function (PDF) and the dependence of the Mean Square Displacement (MSD) of the particles on time. The dependence of the system’s potential energy per particle (PEpP) on time was used to monitor if the system reached the equilibrium state. The PDF of a liquid does not have sharp peaks that may correspond to the lattice spacings and the diffusion rate is very significant in comparison to a crystal state in which diffusion is nearly absent.

2) Then the temperature of the liquid was reduced to some lower value. Sometimes we used an abrupt decrease in temperature (an instant drop) and sometimes we used some cooling rate. For our purposes, the way in which the temperature is reduced is of no significance if the liquid at the reduced temperature remains a liquid with a high diffusion rate. In this case the system reaches its equilibrium state relatively quickly and the cooling history does not influence the properties of the equilibrium state after some relatively short time. We monitored that the equilibrium is reached using the dependence of the PEpP on time. We also monitored that the diffusion rate remains significant and that there do not occur noticeable changes in the PDF with time.

3) When the step 2) is repeated several times relaxation to the equilibrium becomes noticeably slower. Diffusion also slows down. In addition, the lineshape of the PDF starts to exhibit more features that reflect development of some structural ordering. All these changes are well known from simulations of liquids. The point that we would like to make here is that in our simulations crystallization from the liquid state usually happens when some slowness in the relaxation develops. Since our goal was to produce the crystalline state we performed longer simulations of the liquid state at temperatures where slowness in the dynamics is present. This approach usually allowed us to observe crystallization.

4) If we observed crystallization at some temperature then the system was allowed to crystallize and relax for a significant amount of time at this “crystallization” temperature. The relaxation of the system was monitored using the dependence of the PEpP on time. The crystalline states formed in this way can not relax completely and defects in the crystal structures always remains. However, after some time further relaxation becomes quite (very) slow. The PDF of the nearly relaxed state can be calculated and it shows clear qualitative differences with the PDF of the liquid state before crystallization (this is a well known fact that we mention in order to describe how our simulations were performed). It was also observed that diffusion in the crystalline states is nearly absent.

5) On this stage the obtained crystalline state was cooled to nearly zero temperature using some finite cooling rate. This cooling rate was usually quite slow in order to eliminate as many defects as possible. Sometimes, in order to eliminate the defects, we performed longer runs at some fixed temperatures lower than the crystallization temperature. It was found that this approach sometimes indeed helps to produce sharper peaks in the PDFs and correspondingly more pronounced crystal structures. See Ref.[43] for an additional comment.

6) After the crystal structure at the very low temperature was obtained we tried to determine what this crystal structure is. For this we used visual analysis of the crystal structure and the PDF (see section IIB). After we guessed the structure we optimized its parameters in order to minimize the potential energy of the guessed struc-

ture. Then we created the guessed and optimized structure as an input structure file for the LAMMPS program. Of course, this guessed structure does not have any defects. Then we run LAMMPS MD simulations on the guessed and optimized structure at low temperatures. If the guessed structure was stable in the MD simulations then we assumed that our structure-guess might be correct.

7) On this step we heated the guessed crystal structure with some heating rate until its melts. Thus observed “melting” temperature was usually significantly higher than the “crystallization” temperature at the corresponding densities. This heating procedure provides another test for the correctness of the crystal structure guess.

B. Visual analysis of the structures

We performed visual analysis of the structures in two different ways.

One way was to select an atom and consider the geometry of its neighbor environment. Usually we considered several randomly chosen atoms from different regions of the simulation box. Sometimes, when we had the data, we also considered the results from the different runs. In almost all structures that we analyzed in this way we found that the environments of all atoms are similar, if the defects are ignored.

In a different approach we extracted from the whole simulation box some region and then we tried to guess the crystal geometry from the geometry of this region. Usually we considered several extracted regions of different sizes. This helped us to guess the structure and also served as a correctness check for our guess.

III. RESULTS

We performed MD simulations at a number of different densities. In this note we report about the results obtained at the densities $\rho_o\sigma^3 = 2.904$, $\rho_o\sigma^3 = 4.400$, and $\rho_o\sigma^3 = 4.500$. We find these results to be rather unusual.

A. Results for the Density $\rho_o\sigma^3 = 2.904$

We melted the FCC lattice at this density at $T = 0.015$. Then the liquid was cooled using some cooling rate with longer relaxations at lower temperatures when the diffusion is already slow. We clearly observed crystallization, using the dependence of the Potential Energy per Particle (PEpP) on time, at $T \approx 0.0025$. See Fig.1. If thus obtained “crystal” is heated then it melts at $T \approx 0.00438$.

Then the “crystal” obtained through crystallization was further cooled to nearly zero temperature. In or-

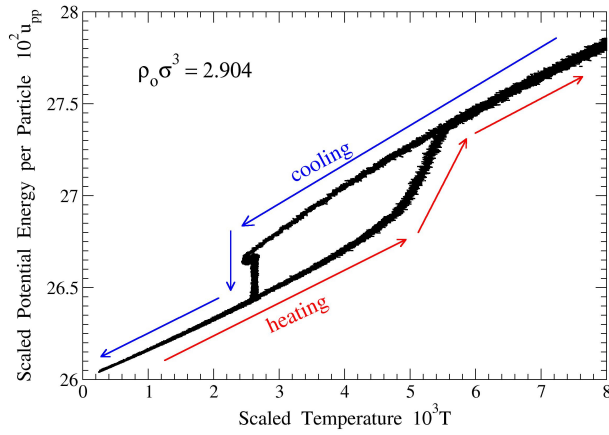


Figure 1. Dependence of the PEPP on temperature combined from the several MD runs at the density $\rho_o \sigma^3 = 2.904$, i.e., at the density at which we observed formation of the “7-columns” rings. In MD simulations we, at first, cooled the system to the temperature $10^3 T = 2.5$. However, at this temperature we were not able to observe crystallization for a rather long time. For this reason we heated the system a bit to the temperature $10^3 T = 2.6$ at which, after some time, we observed crystallization. That is the origin of the “nose” feature around $10^3 T = 2.5$. The shown crystallization-melting hysteresis loop is clearly observable: crystallization happens at $10^3 T = 2.6$, while melting occurs at $10^3 T = 4.6$. Note that the “7-columns” structure remains stable on heating.

der to achieve more relaxed structure we also heated the crystal to the temperatures above $T \approx 0.0025$, but below $T \approx 0.00438$. These trick, however, did not lead to the clearly better relaxed structure. The PDF of the thus obtained structure at nearly zero temperature is shown in Fig.2. Note that the first and second peaks are quite sharp. The third peak is also well pronounced. However, the PDF at larger distances does not look as crystalline in comparison to the previous panels of the same figure.

Further we used visual analysis to examine the “crystal” structure at this density. The results of this analysis are shown in Fig.3,4. We investigated the structure of the “seven-columns rings” in the following way. For a chosen particle in a chosen “seven-columns ring” we looked for its nearest neighbors in the same “seven-columns ring”. We found that usually the chosen particle has two nearest neighbors located at distances $\approx 0.7 \div 0.8$. We also found that usually the chosen particle and its two nearest neighbors are approximately aligned. After all, we found that it is reasonable to assume that every column is formed by 3 spirals and that every spiral is formed, in a chain fashion, by a particle and its 2 nearest neighbors. This geometry is illustrated in Fig. 4.

As far as we understand, the “7-columns” structure of the rings precludes formation of a crystal structure in the plane orthogonal to the axis of the “7-columns” rings. Thus, it is possible that the ground state organization of the columns in the plane orthogonal to the axis of the

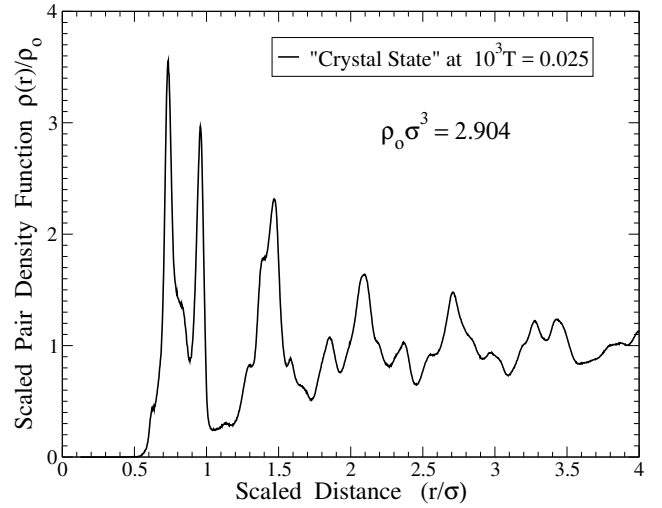


Figure 2. The scaled pair density function, $\rho(r)/\rho_o$, of the structure obtained through crystallization from the liquid state at $\rho_o \sigma^3 = 2.904$ and after cooling to nearly zero temperature.

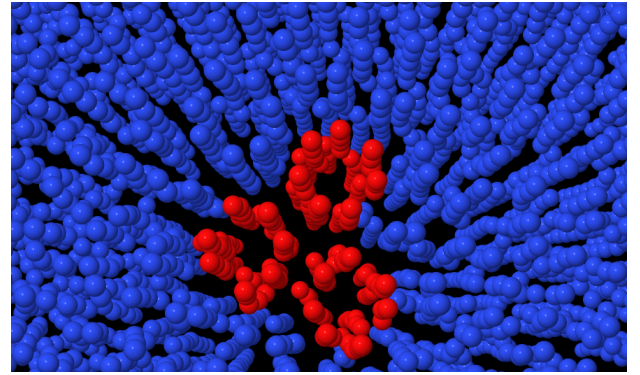


Figure 3. A view on the “crystal” structure at zero temperature at $\rho_o \sigma^3 = 2.904$. The red particles are the same as blue one. The red particles will be used for the further analysis. Note the presence of columns of the particles. Note that some of these columns organize into the rings formed by 7 columns.

columns is quasicrystalline.

In this note we do not discuss the “crystal” structure at this density further.

B. Densities $\rho_o \sigma^3 = 4.40$ and $\rho_o \sigma^3 = 4.50$

At these densities we did not observe crystallization despite rather long molecular dynamics runs at a number of temperatures where the dynamics slows down and becomes very slow. See Fig.5. The PDFs at these densities at very low temperatures are shown in Fig.6(c,d).

We may present more results on simulations at these densities in a separate publication.

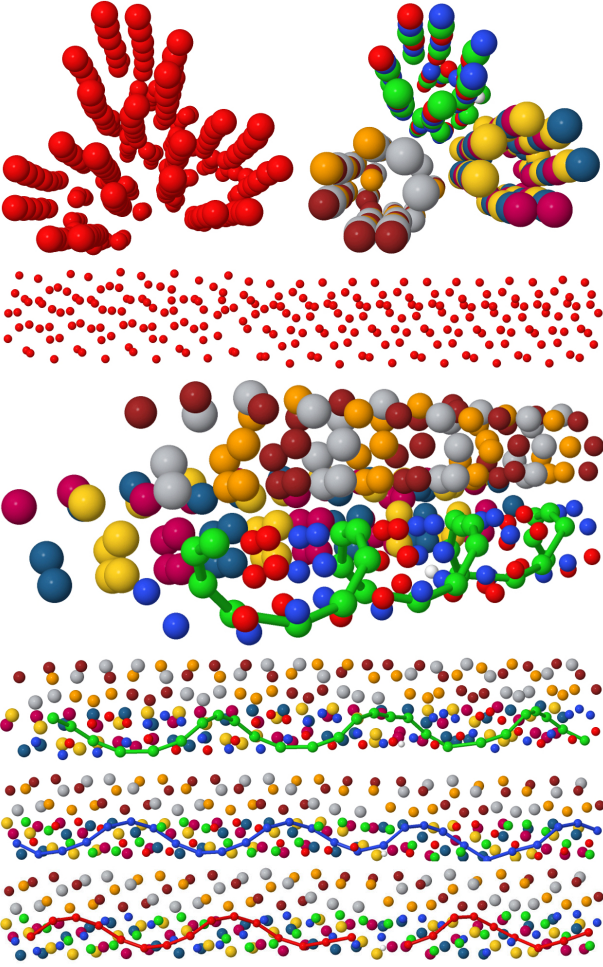


Figure 4. All panels show the same three “7-columns” rings extracted from the structure shown in Fig.3, i.e., those particles which are red in Fig.3. In some panels particles are painted in different colors in order to demonstrate the spiral structure of the “7-columns” rings. We found that every “7-columns” ring can be viewed as consisting of three spirals. For a given particle in the “7-columns” ring its two nearest neighbors belong to the same spiral. The purpose of all panels is to demonstrate the geometry of this organization.

IV. CONCLUSIONS

We investigated behavior of the particles interacting through the harmonic-repulsive potential at different number densities using direct MD simulations. In this note we presented the results obtained for the densities $\rho_o\sigma^3 = 2.904$, $\rho_o\sigma^3 = 4.400$, and $\rho_o\sigma^3 = 4.500$.

1) At the density $\rho_o\sigma^3 = 2.904$ we observed crystallization from the liquid state into a structure in which particles organize into the columns, where each column is formed by 7 (seven) linear chains formed by individual particles. Alternatively one can think that each column is formed by 3 (three) helical coils such that the full circle of each coil involves 7 (seven) particles. Since each column is formed by 7 (seven) linear chains, it is possible,

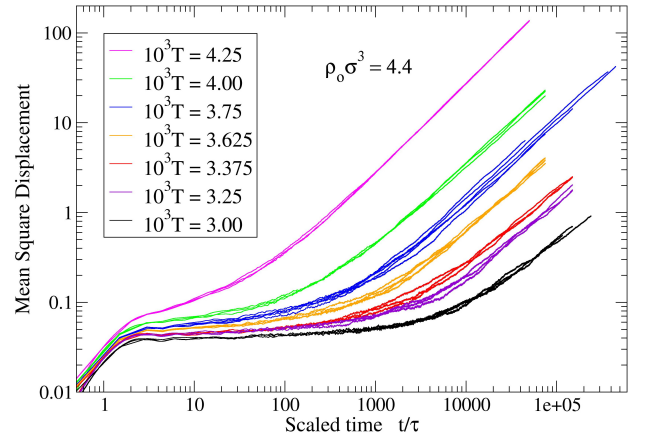


Figure 5. Dependencies of the mean square particles displacement on time in MD runs at the selected temperatures. All data were obtained at the density $\rho_o\sigma^3 = 4.4$. Different curves of the same color correspond to the several consecutive runs at the same temperature. The system at $10^3 T = 3.75$ exhibits very slow relaxation (especially in the earlier runs). This relaxation is partially responsible for the observable differences in the blue curves.

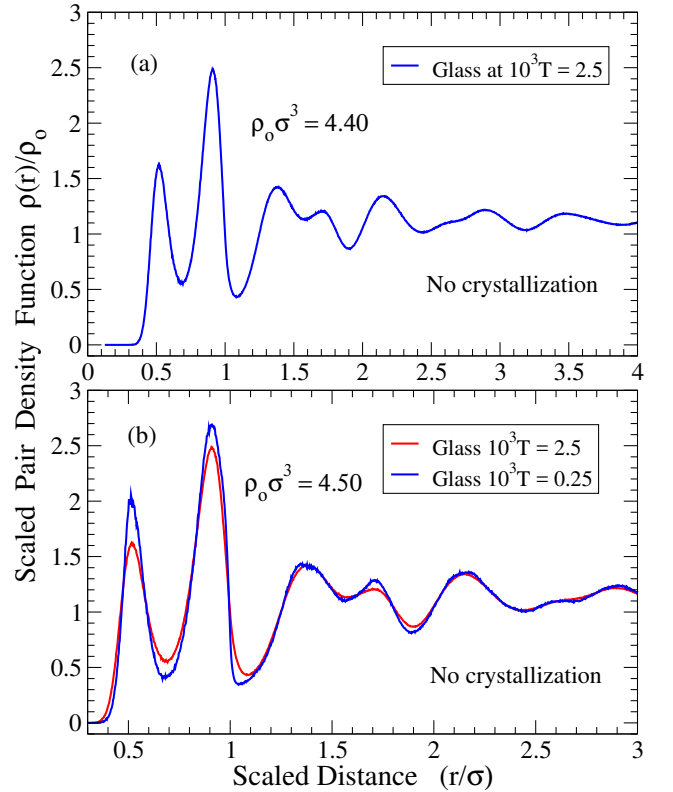


Figure 6. The PDFs, $\rho(r)/\rho_o$, of the glass structures at obtained from the liquid states at densities $\rho_o\sigma^3 = 4.40$ and $\rho_o\sigma^3 = 4.50$. At these densities we did not observe crystallization in the supercooled liquids states despite rather long MD runs (see Fig.5).

in our view, that the formed structure is a quasicrystal.

3) At the densities $\rho_o\sigma^3 = 4.400$ and $\rho_o\sigma^3 = 4.400$ we were not able to observe crystallization despite careful investigations at a number of different temperatures in rather long simulation runs. This result is of interest because this behavior was observed in the single component system (usually single component systems easily crystallize at significant supercooling).

4) At a number of other densities our results (these results are not presented in this note) appear to be in disagreement with the previously predicted phase diagram for the harmonic-repulsive potential [34]. In our view, the reason for the disagreement is that investigations in Ref.[34] were based on the considerations of a certain set of the possible crystal structures (relatively wide and a reasonable set). However, this set did not include many structures that we observed in our simulations. Thus,

in our view, the nature essentially outwitted the initial guess of the possible structures.

The results presented in this note open several obvious routes for further investigations. These include construction of the phase diagram and more detailed investigations of the structural and dynamical properties at the selected values of the densities. Another possible investigation involves direct MD simulations in the NPT ensemble instead of the NVT ensemble used in the current study.

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 - [39] Our goal was to investigate correlations between the eigenvalues of the atomic level stress matrices. In particular, the similarity in the probability distributions of λ_2/λ_1 and λ_3/λ_2 , where $\lambda_1, \lambda_2, \lambda_3$ are the eigenvalues of the atomic level stress matrices.
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 - [43] Sometimes in order to produce a better quality crystalline states we also heated the crystals formed from the liquids above the observed “crystallization” temperatures. Usually we were able to heat the crystals to the temperatures significantly above the observed “crystallization” temperatures. However, this trick combined with the consequent cooling to near zero temperature did not usually lead to the crystal structures which were significantly better than the crystal structures obtained without after-crystallization heating.